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Attention: DO/EO/US

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of

NG, Ka Lok et al

New U.S. National Stage Application of International Application No.

PCT/GB99/01912

International Filing Date:

16 June 1998

For:

THE OPTIMISATION OF GAS FLOW IN REACTORS FOR THE TREAT-

MENT OF GASEOUS MEDIA

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents Washington, D.C. 20231

Sir:

Please amend as follows:

In the Claims:

Cancel all claims presently on file, and substitute therefor the following new claims:

A reactor for the treatment of a gaseous medium, including a cylindrical reactor chamber having an inlet port and an outlet port for a gaseous medium to be processed, a hollow cylindrical gas permeable bed contained within the reactor chamber and substantially co-axial therewith, the gas permeable bed comprising a catalytically active material for interacting with the gaseous medium to promote chemical reaction therein, an annular space between the outside of the bed of active material and the inside of the reactor chamber and means for constraining the gaseous medium to enter the said annular space at one end in an axial direction, the other end of said annular space being closed to axial flow of gaseous medium therefrom, the gaseous medium passing radially through the bed of active material, the said annular space being configured to provide an impedance to the flow of the gaseous medium which increases along

the length of the said annular space in the direction from the said one end towards the said other end.

- 11. A reactor according to claim 10, wherein the width of the said annular space decreases continuously along the length of the said annular space.
- 12. A reactor according to claim 10, wherein there is at least one discontinuous decrease in the width of the said annular space along the length of the said annular space.
- 13. A reactor according to claim 12, wherein there is a single discontinuous decrease in the width of the said annular space approximately at the middle of the said annular space.
- 14. A reactor according to claim 12, wherein there are two discontinuous decreases in the width of the said annular space.
- 15. A reactor according to claim 14, wherein the first discontinuous decrease in the width of the said annular space occurs approximately at the middle of the said annular space and the second discontinuous decrease in the width of the annular space occurs approximately three quarters along the length of the said annular space.
- 16. A reactor according to claim 14, wherein the second discontinuous decrease in the width of the said annular space is less than the first discontinuous decrease in the width of the said annular space.
- 17. A reactor according to claim 10, wherein a first portion of the reactor chamber is provided with at least one axially extending expansion chamber.
- 18. A reactor according to claim 10, wherein the bed of active material is contained between an inner gas permeable elec-

trode and an outer gas permeable electrode co-axial with the said inner electrode and two unpermeable transverse insulating supports, the transverse support nearer the inlet port to the reactor having a plurality of axially directed gas flow passages disposed around its periphery, the transverse support nearer the outlet port of the reactor having a central hole the diameter of which is approximately equal to the diameter of the inner co-axial electrode and the said electrodes and insulating supports are adapted for application between the inner electrode and the outer electrode of a potential sufficient to excite and maintain a plasma in a gaseous medium passing through the bed of active material.

REMARKS

By this Preliminary Amendment, a new set of claims is presented for examination.

Favorable action is courteously solicited.

Respectfully submitted,

William H. Holt Reg. No. 20766

Law Offices of William H. Holt Unit 2, First Floor 1423 Powhatan Street Alexandria, VA 22314

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December **6**, 2000

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The Optimisation of Gas Flow in Reactors for the Treatment of Gaseous Media

The present invention relates to reactors for the treatment of gaseous media and, more specifically to reactors for the removal of noxious substances from the exhaust gases from internal combustion engines.

One type of reactor for the treatment of gaseous

10 media consists of a cylindrical reactor chamber which has inlet and outlet ports by means of which it can be connected into a gas flow system. Inside the reactor chamber, and co-axial within it, is a hollow cylindrical gas permeable bed of active material. By the term "active material", as used herein, is meant a material which interacts with a gaseous medium to promote reaction therein as the gaseous medium passes through the reactor.

A reactor of this type is shown in EP 0608619 (see 20 Figure 2). In a different configuration of reactor for through flow of gaseous media, entering at one end and exiting at the other, the bed of active material is held in place by two supporting disks made of an impermeable material. One support disk has a ring of axially 25 directed holes around its periphery and the other disk has a central hole the diameter of which is approximately equal to the inside diameter of the cylindrical bed of active material. In use a gaseous medium to be processed is admitted to the reactor chamber via the port closer to 30 the first support disk. The gaseous medium is then directed into the annular space between the outside of the cylindrical bed of active material and the wall of the reactor chamber. The closure of this space by the other support disk constrains the gaseous medium to pass 35 radially through the bed of active material prior to leaving the reactor via the central electrode.

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support disks are made of a temperature resistant insulating material and there is provided an electrical connection to the inner electrode by means of which a potential of some kilovolts can be applied to the inner electrode so as to establish a plasma discharge in the gaseous medium in the interstices in the gas permeable bed of active material.

In practice, it has been found that the gas flow

distribution through the bed of active material of such a reactor is uneven, being greater at the downstream end of the bed of active material. Thus the reactor may not operate at its maximum efficiency because the upstream end of the bed of active material may be underused while the downstream end of the bed of active material may be subjected to a higher rate of gas flow than it can usefully process.

It is an object of the present invention to provide 20 an improved reactor of the type described above for the processing of a gaseous medium.

According to the present invention there is provided a reactor for the treatment of a gaseous medium,

25 including a cylindrical reactor chamber having an inlet port and an outlet port for a gaseous medium to be processed, a hollow cylindrical gas permeable bed contained within the reactor chamber and substantially co-axial therewith, the gas permeable bed comprising

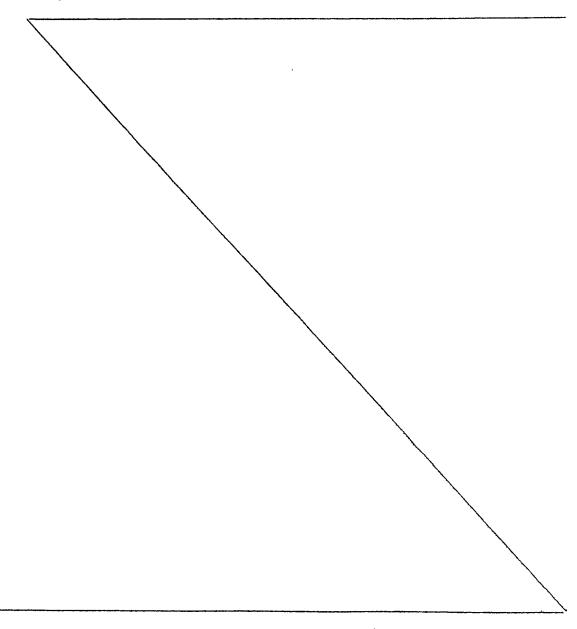
30 catalytically active material for interacting with the gaseous medium to promote chemical reaction therein, an annular space between the outside of the bed of active material and the inside of the reactor chamber and means for constraining the gaseous medium to enter the said

35 annular space at one end in an axial direction, the other end of said annular space being closed to axial flow of

- 2a -

gaseous medium therefrom, the gaseous medium passing radially through the bed of active material, wherein the said annular space is configured to provide an impedance to the flow of the gaseous medium which increases along the length of the said annular space in the direction from the said one end towards the said other end.

The increasing impedance to the axial flow of the gaseous medium through the said annular space preferably 10 is provided by progressively reducing the cross-sectional



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area of the said annular space. The reduction in the cross-sectional area of the said annular space may be continuous, but preferably is discontinuous. A preferred arrangement has two step reductions in the cross-5 sectional area of the said annular space, the first being greater than the second.

According to the present invention in a particular aspect the reactor is for the plasma-assisted treatment of gaseous media, the bed of active material is contained between two gas permeable co-axial disks and two support disks made of an impermeable temperature-resistant insulating material, the support disk nearer the inlet end of the reactor has a plurality of axially directed 15 gas passages around its periphery, and the support disk nearer the outlet end of the reactor has a central hole the diameter of which is substantially equal to the internal diameter of the inner electrode so that a gaseous medium to be processed enters the annular space between the outer electrode and the wall of the reactor chamber axially but is constrained to pass radially through the bed of active material.

Preferably the active material is adapted to remove 25 nitrogenous oxides and carbonaceous combustion products from the exhaust emissions from internal combustion engines.

The invention will now be described, by way of example, 30 with reference to the accompanying drawings, in which:

Figure 1 is a schematic longitudinal section of an existing type of reactor for the plasma-assisted processing of a gaseous medium;

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Figure 2 illustrates how the radial component of gas flow through a bed of active material included in the reactor of Figure 1 varies with distance along the reactor bed from the entrance to the active region of the 5 reactor;

Figure 3 is a schematic longitudinal half-section of a first reactor embodying the invention for the processing of a gaseous medium;

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Figure 4 is a schematic longitudinal half-section of a second reactor embodying the invention for the processing of a gaseous medium;

Figure 5 is a schematic longitudinal half-section of a third reactor embodying the invention for the processing of a gaseous medium;

Figure 6 is a flow diagram showing the radial 20 components of gas flow for the embodiment of Figure 3;

Figure 7 is a flow diagram showing the radial component of gas flow for the embodiment of Figure 4;

Figure 8 is a flow diagram showing the radial component of gas flow for the embodiment of Figure 5; and

Figure 9 is a schematic longitudinal half-section of a fourth embodiment of the invention.

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Referring to Figure 1, a reactor 1 for the plasmaassisted processing of a gaseous medium consists of a stainless chamber 2 which has an inlet stub 3 and an outlet stub 4. The chamber 2 is arranged, in use, to be connected to an earthing point 5. Perforated cylindrical stainless steel electrodes 6 and 7 and positioned co-

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axially within the chamber 2 by means of two impervious electrically-insulating supports 8 and 9. The space 10 bounded by the electrodes 6 and 7 and the insulating supports is filled with a bed 11 of pellets 12 of an active material which has a dielectric constant sufficient to enable a plasma to be established and maintained in the gaseous medium in the interstices between the pellets 12 of the bed 11 of active material. The upstream end 13 of the inner electrode 6 is closed off and arranged to be connected via an insulating feedthrough 14 to a source 15 of an electrical potential sufficient to excite the above-mentioned plasma in the gaseous medium.

The upstream electrode support 8 has a ring of axially-oriented gas passages 16 around its periphery, whereas the downstream electrode support 9 has a central hole 17 in it of approximately the same diameter as the internal diameter of the inner electrode 6. Thus, in use, a gaseous medium to be processed is directed axially into the annular space 18 between the outer electrode 7 and the wall of the chamber 2. As the gas cannot escape from the downstream end of the space 18, it is constrained to enter the bed 11 of active material and pass radially through it.

Figure 2 is a flow diagram showing how the radial component of gas flow for such a reactor varies along the length of the bed of active material. It can be seen that there is very little radial flow through the bed 11 for almost half its length and the radial gas flow increases progressively along the remainder of the bed 11 of active material. Thus, the overall efficiency of the bed 11 of active material is much below that which would be achieved if the radial flow of gas through the bed 11 of the reactor was regular. At present the active

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material at the downstream end of the reactor may be saturated while that at the upstream end of the reactor largely is unused.

5 The present invention seeks to overcome this problem by progressively increasing the resistance to axial flow along the annular space 18 between the wall 2 of the reactor 1 and the outer electrode. Figures 3, 4 and 5 illustrate three ways in which this increase in the resistance to axial flow of the gaseous medium can be achieved.

Unlike Figure 1, these figures are half-sections and extraneous detail has been omitted. However, those parts which are common to all three figures have the same reference numerals.

Referring to Figure 3, a reactor for the processing of a gaseous medium consists of a reactor chamber 300 20 which has inlet and outlet fixing stubs 301 and 302, respectively. Within the reactor chamber 300 is a hollow cylindrical gas permeable bed 303 made of an active material adapted to carry out a desired process on the gaseous medium. For example, the active material may be 25 adapted to catalyse a reaction between one or more components of the gaseous medium. The bed 303 of active materials is contained between two co-axial cylindrical support members 304 and 305, which are gas permeable and two disk transverse supports 306 and 307, made of an 30 unpermeable material, as in the reactor described with reference to Figure 1. If the bed 303 is made of a material which is self supporting, the support members 304 and 305 can be omitted. As before, the support 306 nearer the inlet to the reactor chamber 300 has a number 35 of axially directed gas passages 308 around its periphery and the support 307 nearer the outlet from the reactor

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chamber 300 has a central hole 309 of approximately the same diameter as the inner active bed support member 304. The inner active bed support 304 has a closed, domed end 310 which projects through the support 306 and 5 facilitates the deflection of the incoming gaseous medium towards the periphery of the reactor chamber 300. As with the reactor previously described, a gaseous medium entering the reactor chamber 300 is directed into the annular space 311 between the outer active bed support 305 and the wall of the reactor chamber 300 initially in an axial direction but is then constrained to pass radially through the bed 303 of active material.

However, in the present case, the reactor chamber
300 is not a true cylinder, but tapers in the direction
of gas flow along the space 311 between the outer active
bed support 305 and the wall of the reactor 300. As a
result, the impedance to axial gas flow increases along
the annular space 311 between the outer active bed
support 305 and the wall of the reactor chamber 300, so
increasing the amount of gaseous medium which flows
radially through the upstream part of the bed 303 of
active material. A suitable taper angle is in the region
of two degrees.

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Figure 4 shows a second embodiment of the invention in which the diameter of the reactor chamber 300 is reduced half way along the bed 303 of active material. In a particular example, the width of the annular space 311 between the outer active bed support 305 and the wall of the reactor chamber is 10 mm initially and 5 mm for the second part of the reactor chamber 300. All other components are as for the reactor described with reference to Figure 3.

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Figure 5 shows another embodiment of the invention in which there is a second step-wise reduction in the width of the annular space 311 between the outer active bed support 305 and the wall of the reactor chamber 300 half way along the second part of the bed 303 of active material. In another specific case, the widths of the regions of the annular space 311 between the outer active bed support 305 and the wall of the reactor chamber 300 are 10, 5 and 3 mm.

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Figure 6 is a diagram showing the variation in radial gas flow along the bed 303 of active material for the embodiment of Figure 3. Compared with Figure 2 it can be seen that much more gas passes radially through first half of the bed 303 of active material. In fact the distribution of radial gas flow along the length of the bed 303 of active material is now approximately, symmetrical, but the middle two fifths, approximately of the bed 303 of active material still are underused.

20

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Figure 7 is a diagram showing the variation in radial gas flow along the bed 303 of active material for the embodiment of Figure 4. More gas now passes radially through the first half of the bed 303 of active material than through the second half, with an intermediate peak at the position of the step where the width of the annular space 311 between the outer active bed support 305 and the wall of the reactor chamber 300 is halved. Immediately downstream of this position the radial gas flow rate is less than half that before it. This region of low gas flow extends about one fifth of the length of the bed 303 of active material.

Figure 8 is another gas flow diagram, similar to those of Figures 6 and 7, for the embodiment of Figure 5.

It can be seen that this embodiment of the invention

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gives the most even distribution of radial gas flow through the bed 303 of active material.

Figure 9 shows another embodiment of the invention in which the opposite approach to the problem is adopted. In this embodiment of the invention, instead of reducing the width of the gap 311 between the outer active bed support 305 and the wall of the reactor chamber 300 along the length of the bed 303 of active material so as to cause more gas to be diverted radially in the upstream regions of the bed 303 of active material, a number of axial expansion chambers 901 are provided along the first part of the reactor chamber 300. In this arrangement, not only is the impedance to axial gas flow higher in the 15 second region of the reactor 300 than in the first region, but the expansion chambers 901 reduce the pressure of the gaseous medium as it enters the second region of the reactor chamber 300.

20 The invention has been described above in connection with gas reactors in general. As before, if the reactor is for use in the plasma-assisted processing of gaseous media, specifically, the treatment of the exhaust emissions from internal combustion engines to remove noxious combustion products therefrom, then the inner and 25 outer active bed supports 304 and 305 are made of a metal such as stainless steel and used as electrodes, the outer one being earthed, as is the reactor chamber 300. Also, the transverse supports 306 and 307 have to be made of a temperature resistant insulating material. The material of the active bed 303 has to have a dielectric constant sufficient to enable a plasma to be established and maintained in the interstices within the bed of active material.

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Claims

- A reactor for the treatment of a gaseous medium, including a cylindrical reactor chamber (300) having an inlet port (301) and an outlet port (302) for a gaseous medium to be processed, a hollow cylindrical gas permeable bed (300) contained within the reactor chamber (300) and substantially co-axial therewith, the gas permeable bed comprising a catalytically active material
- for interacting with the gaseous medium to promote chemical reaction therein, an annular space (311) between the outside of the bed of active material (303) and the inside of the reactor chamber (300) and means (306) for constraining the gaseous medium to enter the said annular
- space (311) at one end in an axial direction, the other end of said annular space (311) being closed to axial flow of gaseous medium therefrom, the gaseous medium passing radially through the bed (303) of active material, characterised in that the said annular space
- 20 (311) is configured to provide an impedance to the flow of the gaseous medium which increases along the length of the said annular space (311) in the direction from the said one end towards the said other end.
- 25 2. A reactor according to claim 1, further characterised in that the width of the said annular space (311) decreases continuously along the length of the said annular space (311).
- 30 3. A reactor according to claim 1, further characterised in that there is at least one discontinuous decrease in the width of the said annular space (311) along the length of the said annular space (311).
- 35 4. A reactor according to claim 3, further characterised in that there is a single discontinuous

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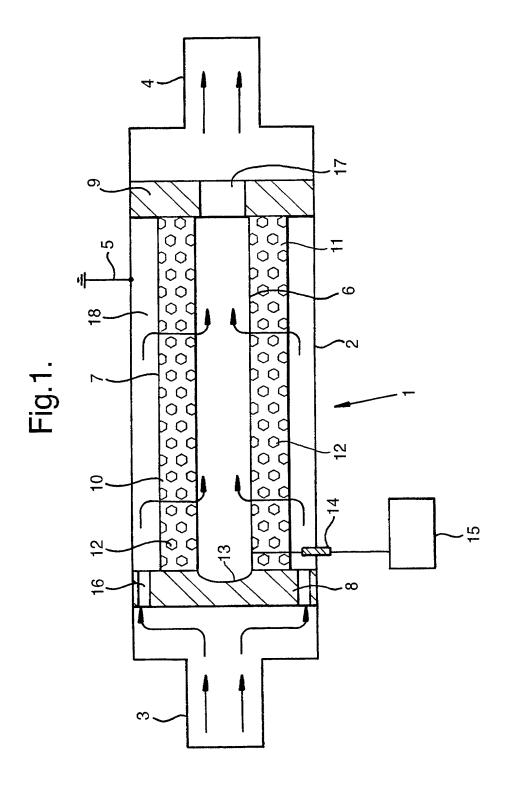
decrease in the width of the said annular space (311) approximately at the middle of the said annular space (311).

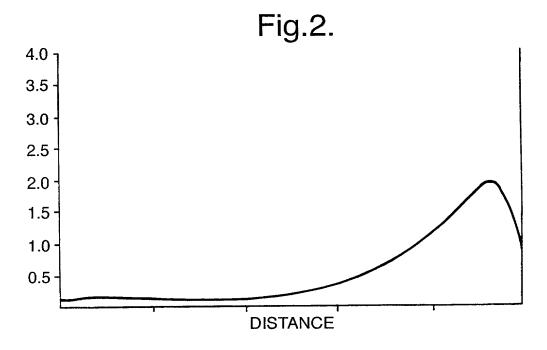
- 5. A reactor according to claim 3, further characterised in that there are two discontinuous decreases in the width of the said annular space (311).
- 6. A reactor according to claim 5, further
 10 characterised in that the first discontinuous decrease in the width of the said annular space (311) occurs approximately at the middle of the said annular space (311) and the second discontinuous decrease in the width of the annular space (311) occurs approximately three quarters along the length of the said annular space (311).
- 7. A reactor according to claim 5, further characterised in that the second discontinuous decrease 20 in the width of the said annular space (311) is less than the first discontinuous decrease in the width of the said annular space (311).
- 8. A reactor according to claim 1, further
 25 characterised in that a first portion of the reactor chamber (300) is provided with at least one axially extending expansion chamber (901).
- 9. A reactor according to any preceding claim, further characterised in that the bed (303) of active material is contained between two co-axial gas permeable electrodes (304, 305) and two unpermeable transverse insulating supports (306, 307), the transverse support (306) nearer the inlet port (301) to the reactor has a plurality of axially directed gas flow passages (308) disposed around
- 35 axially directed gas flow passages (308) disposed around its periphery, the transverse support (307) nearer the

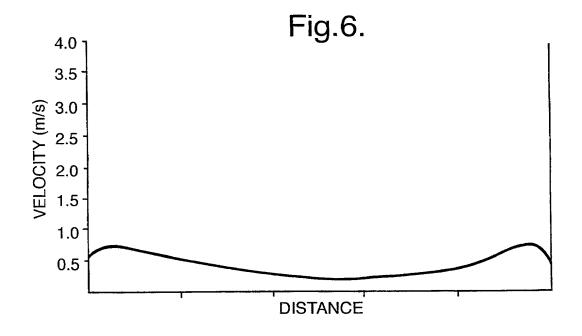
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outlet port (302) of the reactor has a central hole (309) the diameter of which is approximately equal to the diameter of the inner co-axial electrode (304) and there is provided means for applying to the inner electrode (304) a potential sufficient to excite and maintain a plasma in a gaseous medium passing through the bed (303) of active material.

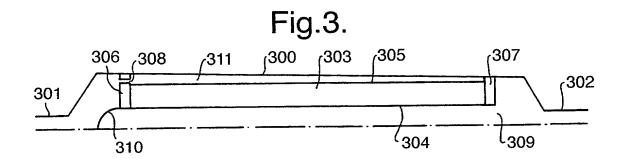
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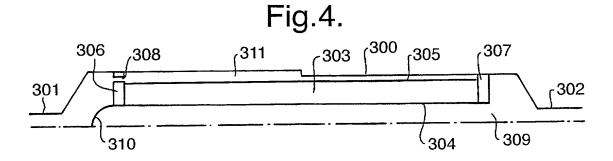


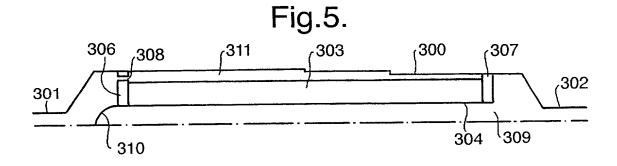


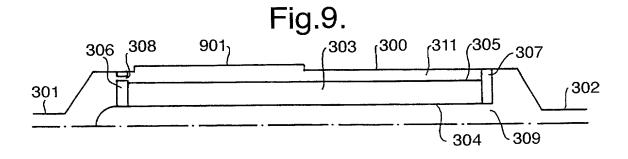


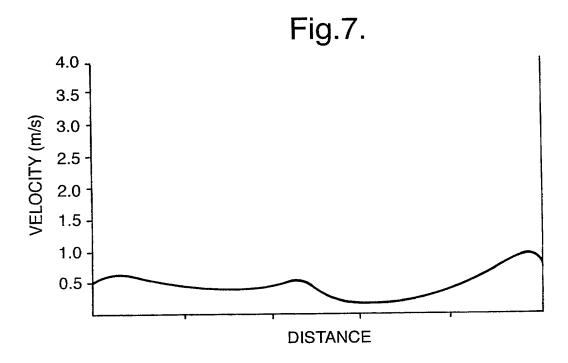
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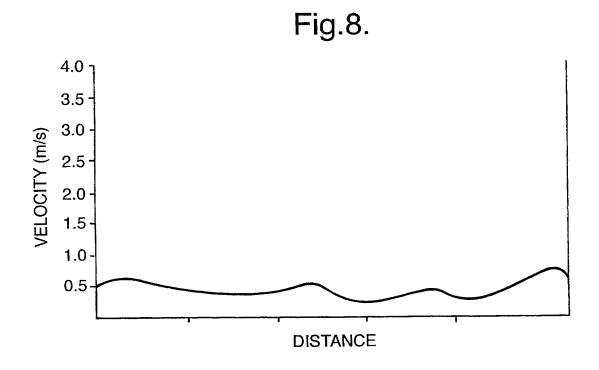












COMBINED DECLARATION AND POWER OF ATTORNEY FOR PATENT APPLICATION (USA)

(INCLUDING DESIGN PATENT APPLICATIONS)

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name. I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled The optimisation of gas flow in reactors for the treatment of gaseous media the specification of which is attached hereto, unless the following box is checked: as United States Application Number or PCT International Application on 9th June 2000 and (if applicab 16th June 1999 was filed on (if applicable). No. PCT/GB99/01912 and was amended on I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, \$1.56. I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed: Priority Claimed 24th June 1998 [X] United Kingdom [] 98 13482.8 Yes Mο (Day/Month/Year Filed) (Country) (Number) Priority Claimed [1 [] (Day/Month/Year Filed) Νo Yes (Country) (Number) [] Additional applications identified on attached sheet. I hereby claim the benefit under Title 35, United States Code, \$120 of any United States application(s) listed below and, insofar as the subject matter of each of the 1 claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application: L Ų. (Status) (patented, pending, abandoned) (Application Serial Number) (Filing Date) TI: (Status) (patented, pending, abandoned) ű. (Filing Date) (Application Serial Number) [] Additional applications identified on attached sheet. I hereby appoint the following attorneys to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith: Howard M. Ellis, Reg. No. 25856 Ronald E. Greigg, Reg. No. 31517; D. Peter Hochberg, Reg. No. 24603; William H. Holt, Reg. No. 20766; WILLIAM H. HOLT, Telephone Number: (703) 838-2700 Direct all telephone calls to LAW OFFICES OF WILLIAM H. HOLT, 1423 Powhatan Street, Unit 2, First Floor, Alexandria, Virginia 22314... Address all correspondence to I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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